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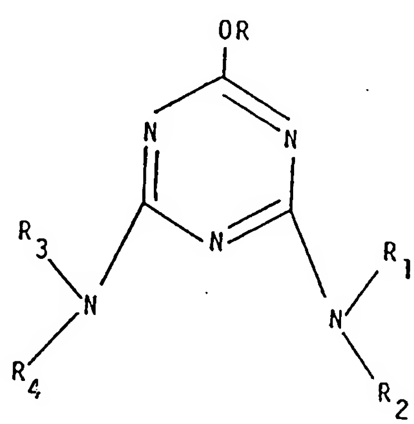
(54) **Self-extinguishing polymeric compositions.**

(57) Described are self-extinguishing polymeric compositions comprising:

a) from 91 to 40 parts by weight of thermoplastic polymer or of polymer having elastomeric properties;

b) from 6 to 33 parts by weight of one or more ammonium and/or amine phosphates and/or phosphonates;

c) from 3 to 27 parts by weight of one or more 2,4-diamino-6-hydroxy-1,3,5-triazine derivatives of general formula (I):



(I)

EP 0 475 368 A2

wherein R, R_1, R_2, R_3 and R_4 are defined as stated in claim 1.

The present invention relates to self-extinguishing polymeric compositions based on thermoplastic polymers and/or polymers endowed with elastomeric properties, especially olefinic polymers and copolymers, which compositions comprise specific triazine derivatives in combination with ammoniun and/or amine phosphates and/or ammonium and/or amine phosphonates.

Several methods of reducing or eliminating the combustibility of polymers are known in the art. Some of said methods are based on the use of metal compounds, in particular on antimony, bismuth or arsenic, in combination with partially halogenated, thermally unstable organic compounds, such as chlorinated paraffin waxes.

Other methods are based on the use of substances capable of yielding intumescence. The formulations of the intumescent type generally are composed of the polymer and at least three main additives, i.e., an essentially phosphorus-containing additive, whose purpose is to form, during the combustion, an impermeable, semi-solid and vitreous layer essentially composed of polyphosphoric acid and to activate the process of formation of intumescence; a second additive, containing nitrogen, which is to serve as foaming agent; and a third, carbon-containing additive, which acts as a carbon donor in order to allow an insulting cellular carbonaceous layer ("char") to be formed between the polymer and the flame.

Examples of intumescent formulations of said type may be found in US-A-3,810,862 (Phillips Petroleum Co.; based on melamine, pentaerythritol and ammonium polyphosphate); US-A-4,727,102 (Vamp S.r.l.; based on melamine cyanurate, a hydroxyalkyl derivate of isocyanuric acid and ammonium polyphosphate); and WO-85/05626 (Plascoat U.K. Limited; on the basis of various phosphorus and nitrogen compounds among which, in particular, a combination of melamine phosphate, pentaerythritol and ammonium polyphosphate may be mentioned).

In more recent formulations, together with an organic or inorganic phosphorus compound, a nitrogen-containing organic compound was used, generally consisting of an aminoplastic resin obtained by means of condensation of urea, melamine or dicyandiamide with formaldehyde.

Examples of formulations containing two additives are those described in US-A-4,504,610 (Montedison S.p.A.; based on oligomeric derivatives of 1,3,5-triazine and ammonium polyphosphate) and EP-A-14,463 (Montedison S.p.A.; based on organic compounds selected from benzylguanamine and reaction products of aldehydes and several cyclic nitrogen compounds, in particular benzylguanamine-formaldehyde copolymers, and ammonium polyphosphate).

Self-extinguishing compositions can also be obtained by using single-component additives, which contain in their organic molecule both nitrogen and phosphorus atoms, as disclosed in US-A-4,201,705 (Borg-Warner Corp.).

These intumescent flame retardant systems endow the polymers to which they are added with the property of forming a carbonaceous residue when they burn or are exposed to a flame. This kind of flame-retardant system offers numerous advantages, i.e., absence of corrosion phenomena in the machinery in which the polymers are processed, a lower emission of smokes as compared to systems containing metal compounds and halogenated hydrocarbons, and, above all, the possibility of endowing the polymers with satisfactory flame-proof properties with a smaller amount of total additive and therefore without excessively impairing the mechanical properties thereof.

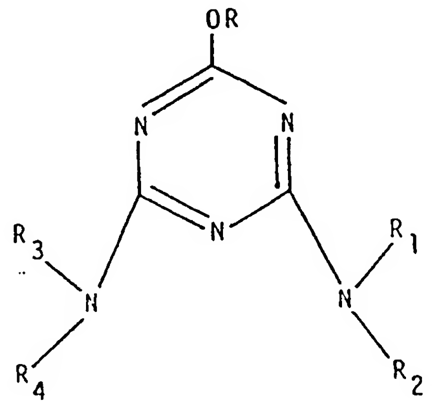
It has now surprisingly been found that polymeric compositions showing very good anti-flame properties may be obtained by adding a certain class of 2,4-diamino-6-hydroxy-1,3-5-triazine derivatives to the corresponding polymers, the effectiveness of said derivatives being higher than that of the products known from the prior art.

More specifically the compositions of the present invention comprise:

a) from 91 to 40, preferably 88 to 60, parts by weight of thermoplastic polymer and/or of polymer having elastomeric properties;

b) from 6 to 33, preferably 8 to 30, parts by weight of one or more ammonium and/or amine phosphates and/or phosphonates;

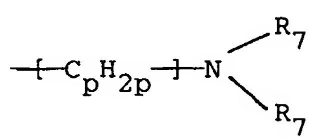
c) from 3 to 27, preferably 4 to 20, parts by weight of one or more compounds derived from 2,4-diamino-6-hydroxy-1,3,5-triazine and having the general formula (I):



(I)

wherein:

R = H; $(C_2\text{-}C_6)$-alkenyl; $(C_6\text{-}C_{12})$-cycloalkyl; $(C_6\text{-}C_{12})$-alkylcycloalkyl; $(C_6\text{-}C_{12})$-aryl; $(C_7\text{-}C_{12})$-aralkyl; and $\{C_nH_{2n}\}X$, n being an integer of from 1 to 8, preferably from 1 to 4 and X being selected from H; CN; -O-$(C_1\text{-}C_4)$-alkyl;-O-$(C_2\text{-}C_4)$-alkenyl; $(C_6\text{-}C_{12})$-cycloalkyl; $(C_6\text{-}C_{12})$-alkylcycloalkyl;-O-$(C_6\text{-}C_{12})$-aryl; moieties -N-$(R_5)_2$ wherein the groups R_5, the same or different from each other, represent $(C_1\text{-}C_4)$-alkyl or $(C_3\text{-}C_4)$-alkenyl; or the moiety -$N(R_5)_2$ is replaced by an N-heterocyclic radical which optionally contains another heteroatom (preferably selected from O, N and S) and is linked to the alkyl chain through the nitrogen atom; (X being preferably H); at least one of the groups R_1, R_2, R_3 and R_4 is selected from: $\{C_mH_{2m}\}O\text{-}R_6$;



wherein

m = an integer of from 2 to 8, preferably from 2 to 4;

p = an integer of from 2 to 6;

R_6 = H; $(C_1\text{-}C_8)$-, preferably $(C_1\text{-}C_4)$-alkyl; $(C_2\text{-}C_6)$-alkenyl; $\{C_qH_{2q}\}O\text{-}R_8$, wherein q is an integer of from 1 to 4 and R_8 is hydrogen or $(C_1\text{-}C_4)$-alkyl; $(C_6\text{-}C_{12})$-cycloalkyl or $(C_6\text{-}C_{12})$-alkylcycloalkyl;

the groups R_7, the same or different from each other, represent H; $(C_1\text{-}C_8)$-alkyl; $(C_2\text{-}C_6)$-alkenyl; $(C_1\text{-}C_4)$-hydroxyalkyl; $(C_6\text{-}C_{12})$-cycloalkyl and $(C_6\text{-}C_{12})$-alkylcycloalkyl;

or the moiety -$N(R_7)_2$ is replaced by an N-heterocyclic radical which optionally contains another heteroatom (preferably selected from O, N and S) and is linked to the alkyl chain through the nitrogen atom;

or in general formula (I) at least one of the moieties -NR_1R_2 and -NR_3R_4 is replaced by a N-heterocyclic radical which optionally contains another heteroatom (preferably selected from O, N and S) and is linked to the triazine ring through the nitrogen atom;

the remaining groups R_1 to R_4, the same or different from each other, having the meanings mentioned above or being selected from H; $(C_1\text{-}C_{18})$-alkyl; $(C_2\text{-}C_8)$-alkenyl; $(C_6\text{-}C_{16})$-cycloalkyl and $(C_6\text{-}C_{16})$-alkylcycloalkyl, optionally substituted by a hydroxy or$(C_1\text{-}C_4)$-hydroxyalkyl group.

The above compounds of general formula (I), besides having a simple structure and being obtainable by attaching a suitable substituent selected from those described hereinafter to 2,4-diamino-6-hydroxy-1,3,5-triazine which as such is unable to endow the above polymers with self-extinguishing properties are particularly heat-stable and therefore maintain a high flame-retardant activity also after a heat processing of the polymeric compositions containing them.

Furthermore, the compositions of the present invention show the advantage of resulting, in the event of a fire, in a very low and not darkening smoke emission.

Specific examples of the group R in general formula (I) are methyl; ethyl; propyl; isopropyl; n-butyl; isobutyl; tert-butyl; n-pentyl; isopentyl; n-hexyl; tert-hexyl; octyl; tert-octyl; ethenyl; propenyl; butenyl; isobutenyl; hexenyl; cyclohexyl; propylcyclohexyl; butylcyclohexyl; phenyl; benzyl; 2-phenyl-ethyl; cyanomethyl; 2-cyanoethyl; 2-methoxyethyl; 2-methoxypropyl; 3-methoxypropyl; 4-methoxybutyl; 5-methoxypentyl; 6-methoxyhexyl; 7-methoxyheptyl; 7-methoxyoctyl; 2-ethoxyethyl; 3-ethoxypropyl; 4-ethoxybutyl; 5-

ethoxypentyl; 2-cyclohexyloxyethyl; 2-ethenyloxyethyl; 2-phenoxyethyl; 2-(N,N-dimethylamino)ethyl; 3-(N,N-dimethylamino)propyl; 4-(N,N-dimethylamino)butyl; 5-(N,N-dimethylamino)pentyl; 6-(N,N-dimethylamino)-hexyl; 2-(N,N-diethylamino)ethyl; 3-(N,N-diethylamino)propyl; 4-(N,N-diethylamino)butyl; 5-(N,N-diethylamino)pentyl; 2-(N,N-dipropylamino)ethyl; 2-(N-methyl-N-1-propenylamino)ethyl; 2-(N,N-di-1-propenylamino)ethyl; 4-(N,N-di-1-propenylamino)butyl;etc.

Specific examples of groups R_1 to R_4 in general formula (I) are methyl; ethyl; propyl, isopropyl; n-butyl; isobutyl; tert-butyl; n-pentyl; isopentyl; n-hexyl; tert-hexyl; octyl; tert-octyl; decyl; dodecyl; octadecyl; ethenyl; propenyl; butenyl; isobutenyl; hexenyl; octenyl; cyclohexyl; propylcyclohexyl; butylcyclohexyl; decylcyclohexyl; hydroxycyclohexyl; hydroxyethylcyclohexyl; 2-hydroxyethyl; 2-hydroxypropyl; 3-hydroxypropyl; 3-hydroxybutyl; 4-hydroxybutyl; 3-hydroxypentyl; 5-hydroxypentyl; 6-hydroxyhexyl; 3-hydroxy-2,5-dimethylhexyl; 7-hydroxyheptyl; 7-hydroxyoctyl; 2-methoxyethyl; 2-methoxypropyl; 3-methoxypropyl; 4-methoxybutyl; 6-methoxyhexyl; 7-methoxyheptyl; 7-methoxyoctyl; 2-ethoxyethyl; 3-ethoxypropyl; 4-ethoxybutyl; 3-propoxypropyl; 3-butoxypropyl; 4-butoxybutyl; 4-isobutoxybutyl; 5-propoxypentyl; 2-cyclohexyloxyethyl; 2-ethenyloxyethyl; 2-(N,N-dimethylamino)ethyl; 3-(N,N-dimethylamino)propyl; 4-(N,N-dimethylamino)-butyl; 5-(N,N-dimethylamino)pentyl; 4-(N,N-diethylamino)butyl; 5-(N,N-diethylamino)pentyl; 5-(N,N-diisopropylamino)pentyl; 3-(N-ethylamino)-propyl; 4-(N-methylamino)butyl; 4-(N,N-dipropylamino)butyl; 2-(N,N-diisopropylamino)ethyl; 6-(N-hexenylamino)hexyl; 2-(N-ethenylamino)ethyl; 2-(N-cyclohexylamino)ethyl; 2-(N-2-hydroxyethylamino)ethyl; 2-(2-hydroxyethoxy)ethyl; 2-(2-methoxyethoxy)ethyl; 6-(N-propylamino)-hexyl; etc.

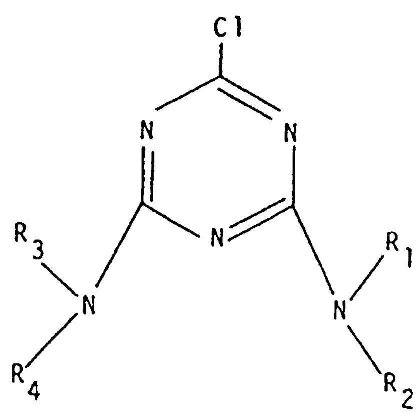
Specific examples of heterocyclic radicals which may replace the moieties $-NR_1R_2$ and $-NR_3R_4$ in general formula (I) are aziridyl, pyrrolidyl, piperidyl; morpholyl; thiomorpholyl, piperazyl; 4-methylpiperazyl; 4-ethylpiperazyl; 2-methylpiperazyl; 2,5-dimethylpiperazyl; 2,3,5,6-tetramethylpiperazyl; 2,2,5,5-tetramethylpiperazyl; 2-ethylpiperazyl; 2,5-diethylpiperazyl; etc.

Specific examples of heterocyclic radicals which can replace the moiety $-N(R_5)_2$ are pyrrolidyl; piperidyl; morpholyl; thiomorpholyl; piperazyl; 4-methylpiperazyl; 4-ethylpiperazyl; etc.

Specific examples of heterocyclic radicals which can replace the moiety $-N(R_7)_2$ are aziridyl; pyrrolidyl; piperidyl; morpholyl; thiomorpholyl; piperazyl; 4-methylpiperazyl; 4-ethylpiperazyl; etc.

Particularly preferred are the compounds of general formula (I) wherein R is hydrogen.

Compounds of general formula (I) can be prepared from the intermediates of general formula (II):



(II)

wherein R_1 to R_4 have the previously defined meaning, according to the following modalities:

a) when R is hydrogen by hydrolysis, either with an acid (such as hydrochloric acid, hydrobromic acid, sulfuric acid, phosphoric acid, etc.) at temperatures of from about 60 to about 100°C, or with a base (such as sodium hydroxide, potassium hydroxide, etc.), at temperatures of from about 100 to about 180°C;

b) when R is different from hydrogen, by a condensation reaction with a compound of general formula (III):

R-OH (III)

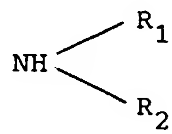
wherein R has the previously defined meanings, in a solvent (such as toluene, xylene, ortho-dichlorobenzene, etc.) and/or in an excess of compound (III) if the latter can act as solvent (e.g., in the case of

methanol, ethanol, etc.) and in the presence of a base (such as sodium hydroxide, potassium hydroxide, sodium metal, etc.) at temperatures of from about 60 to about 150 °C.

The product formed can easily be separated from the reaction mass by filtration.

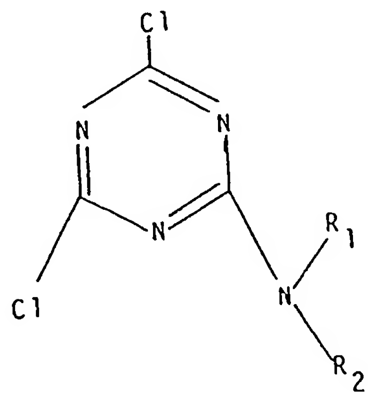
Generally products of general formula (I) of good quality and being in the form of a white crystalline powder are obtained which can be used in the present self-extinguishing compositions without any further purification.

The intermediates of general formula (II) can easily be synthesized by reacting a halide of cyanuric acid, e.g., the chloride, at a temperature of from about 0 to about 10 °C and in a solvent (such as acetone, water, methylene chloride, etc.), with an amine or general formula (IV):



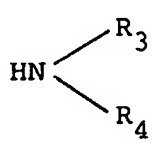
(IV)

wherein R_1 and R_2 have the previously defined meaning. Said reaction may be carried out in the presence or absence (depending on the molar ratio used) of an acidity acceptor (such as NaOH, $NaHCO_3$, Na_2CO_3, triethylamine, etc.) and yields the intermediate of general formula (V):



(V)

This intermediate, either separated or not, is then allowed to react under conditions analogous to the preceding ones, but at a higher temperature, for instance of from about 10 to about 50 °C, with an amine of general formula (VI):

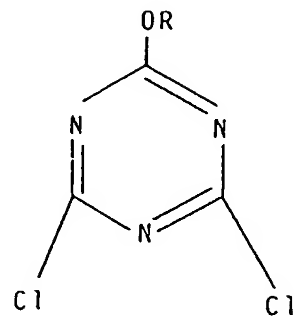


(VI)

wherein R_3 and R_4 have the previously defined meanings.

If it is desired to obtain derivatives of the general formula (I) wherein $-NR_1R_2 = -NR_3R_4$, one mol of the halide of cyanuric acid is allowed to react with two mols (in the presence of an acidity acceptor) or four mols (in the absence of the acidity acceptor) of an amine of general formula (IV) above under conditions analogous to those described previously.

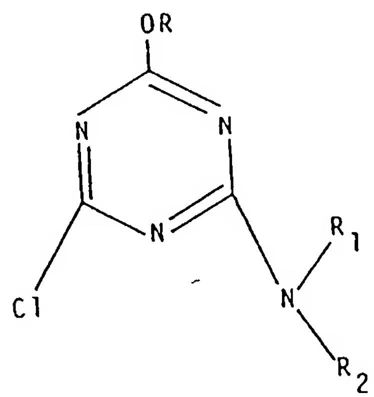
An alternative method for obtaining compounds of general formula (I) wherein R is different from hydrogen, comprises the reaction of a halide of cyanuric acid, e.g., the chloride, with a derivative of general formula (III), at about 10 to about 110 °C in a suitable solvent (such as acetone, methylene chloride, toluene, xylene etc.) and/or in an excess of the reagent of general formula (III) if the latter can act as solvent (as in the case of methanol, ethanol, etc.) and in the presence of an acidity acceptor (such as $NaHCO_3$, Na_2CO_3, NaOH, triethylamine, collidine, etc.), whereby the intermediate of general formula (VII):



(VII)

wherein R has the previously defined meanings, is obtained.

This intermediate, either separated or not, is allowed to react with an amine of general formula (IV) under conditions analogous to the preceding ones, but at temperatures of from about -5 to about 30 °C, to give the intermediate of general formula (VIII):



(VIII)

wherein R, R_1 and R_2 have the previously defined meanings.

This intermediate, either separated or not, is allowed to react again under conditions analogous to the preceding ones, but at a higher temperature (for instance 40 to 120 °C) and therefore with a solvent also compatible with these temperatures (such as water, toluene, xylene, etc.) with an amine of general formula (VI) thus affording compounds of general formula (I).

If compounds of general formula (I) are desired wherein R is different from hydrogen also in this case, and $-NR_1R_2 = -NR_3R_4$, one mole of the intermediate of general formula (VII) is allowed to react with two mols (in the presence of an acidity acceptor) or with four mols (in absence of an acidity acceptor) of an amine of general formula (IV) under conditions analogous to those previously described.

From compounds of general formula (I) wherein R is different from hydrogen (and is preferably (C_1-C_4)-alkyl) it is possible to obtain derivatives of general formula (I) wherein R is hydrogen, by hydrolysis reaction either with an acid (working at tempertaures of from about 80 to about 140 °C) or with a base (working at temperatures of from about 100 to about 180 °C), using the same reagents as given for the hydrolysis of intermediates of general formula (II). Among the phosphates, ammonium polyphosphates of the general formula

$(NH_4)_{n+2}P_nO_{3n+1}$

wherein n is an integer equal to, or greater than, 2, are preferred. The molecular weight of the polyphosphates should be high enough to secure a low solubility in water.

For exemplary purposes, n preferably is comprised within the range of from 2 to 500.

The composition of the polyphosphates having the formula indicated hereinabove, in which n is a high enough number, and preferably ranges from 5 to 500, practically corresponds to a metaphosphate of formula

$(NH_4PO_3)_n$.

An example of said polyphosphates is the product known under the trade name "Exolit ® 422" (Hoechst), having the composition $(NH_4PO_3)_n$ in which n is greater than 50. Another example is the product known under the name "Phos-Chek ® P/30" (Monsanto Chemical) which has a similar composition.

Another polyphosphate which can be used advantageously, above all thanks to its low solubility in water, is the product known under the trade name "Exolit ® 462" (Hoechst) which is "Exolit ® 422" microencapsulated in melamine-formaldehyde resin.

Other phosphates which can be used are those which are derived from amines, such as, e.g., dimethyl ammonium phosphate or diethyl ammonium phosphate, ethylene diamine phosphate, melamine ortho-phosphate or melamine pyrophosphate.

Good results may also be obtained by using mono- or polyammonium phosphonates selected from the salts derived from mono-or polyphosphonic acids.

Examples of said acids are:
ethane-1,1,2-triphosphonic acid, 2-hydroxy-ethane-1,1,2-triphosphonic acid, propane-1,2,3-triphosphonic acid, methylphosphonic acid, ethylphosphonic acid, n-propylphosphonic acid, n-butylphosphonic acid, phenylphosphonic acid, 1-amino-ethane-1,1-diphosphonic acid, 1-hydroxy-ethane-1,1-diphosphonic acid, 1-hydroxy-dodecane-1,1-diphosphonic acid, phosphonoacetic acid, 2-phosphonopropionic acid, 3-phosphonopropionic acid, 2-phosphonobutyric acid, 4-phosphonobutyric acid, amine tri-(methylenephosphonic) acid, ethylenediamine tetra(methylenephosphonic) acid, hexamethylene diamine tetra(methylenephosphonic) acid, diethylene triamine penta(methylenephosphonic) acid, etc.

Among the polymers which can be used in the compositions according to the present invention, preferred are polymers and copolymers of olefins of general formula

$$R' - CH = CH_2$$

wherein R' is hydrogen or a (C_{1-8})-alkyl or (C_{6-10})-aryl radical, in particular:

(1) isotactic or prevailingly isotactic polypropylene;

(2) HDPE, LLDPE and LDPE polyethylene;

(3) crystalline copolymers of propylene and minor amounts of ethylene and/or of other alpha-olefins, such as, e.g., 1-butene, 1-hexene, 1-octene, 4-methyl-1-pentene;

(4) heterophasic compositions comprising (A) a fraction constituted by a propylene homopolymer, or by one of the copolymers defined under (3); and (B) a copolymeric fraction constituted by elastomeric copolymers of ethylene and an alpha-olefin, optionally containing minor proportions of one or more dienes, wherein the alpha-olefin is preferably selected from propylene and 1-butene; and

(5) elastomeric copolymers of ethylene and alpha-olefin(s), optionally containing minor proportions of one or more dienes.

Examples of dienes more commonly contained in said elastomeric copolymers are butadiene, ethylidene-norbornene and hexadiene-1,4.

Among the polymers of olefins of general formula $R'\text{-}CH=CH_2$ wherein R' is an aryl radical, "crystal" and impact-resistant polystyrene are preferred.

Other examples of polymers which can be used are acrylonitrile/butadiene/styrene terpolymers (ABS); styrene/acrylonitrile copolymers (SAN); polyurethanes (of polyester or polyether grade); poly (ethylene terephthalate); poly (butylene terephthalate); polyamides; etc.

The self-extinguishing compositions according to the present invention can be prepared according to methods known in the art. For example, ammonium and/or amine phosphate and/or phosphonate is first intimately mixed with one or more finely ground nitrogen-containing compounds of general formula (I) (the particles of which generally are smaller than 70 μm) and the thus obtained mixture is added to the polymer in a turbo-mixer, in order to generate a homogeneous mixture which is either extruded or granulated. The thus obtained granular product can be transformed into various finished articles according to any of the well-known moulding techniques.

The fire-retardant additives according to the present invention are also suitable for use in the field of flame-retardant paints.

Compounds comprised in the general formula (I), besides those exemplified, which can advantageously be used in the polymeric compositions of the present invention are those listed in Table 1 below.

TABLE 1

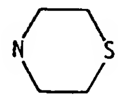
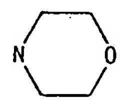
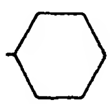
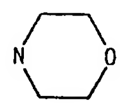
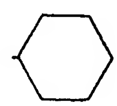
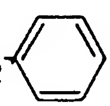
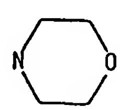
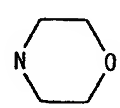
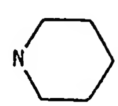
Compound No.	R	R_1—(N)—R_2	R_3—(N)—R_4
1	H		H H
2	C_2H_5	$CH_2\underset{CH_3}{\underset{\mid}{C}}HOH$ H	H H
3	CH_3		C_2H_5 H
4	CH_3	CH_2CH_2OH C_2H_5	H H
5			H H
6	H	CH_2CH_2OH H	 H
7	CH_2-	$CH_2CH_2OCH_3$ H	$CH_2CH_2OCH_3$ H
8	CH_2CH_2CN	$CH_2CH_2CH_2OH$ H	$CH_2CH_2CH_2OH$ H
9	CH_3		
10	CH_3		H H

TABLE 1

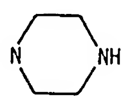
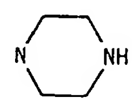
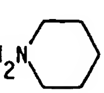
Compound No.	R	R_1 —(—N—)— R_2		R_3 —(—N—)— R_4	
11	H	$(CH_2)_3OC_2H_5$	H	$(CH_2)_3OC_2H_5$	H
12	C_2H_5	$CH_2CH(CH_3)OH$	H	$CH_2CH(CH_3)OH$	H
13	$n\text{-}C_4H_9$	$CH_2CH(CH_3)OH$	H	H	H
14	CH_3	$(CH_2)_3OCH_3$	H	$(CH_2)_3OCH_3$	H
15	$CH_2CH_2N(CH_3)_2$	CH_2CH_2OH	H	CH_2CH_2OH	H
16	H				
17	H	CH_2CH_2OH	CH_2CH_2OH	H	H
18	H	$(CH_2)_4OCH_3$	H	$(CH_2)_4OCH_3$	H
19	C_2H_5	$CH_2CH_2CH_2$	H	H	H
20	CH_3	$CH_2\text{-}C(CH_3)=CH_2$	H	H	H

TABLE 1 (continued)

Compound No.	R	R_1—(—N—)—R_2		R_3—(—N—)—R_4	
21	$CH_2CH_2OCH_3$	CH_2CH_2OH	H	morpholino (N-ring-O)	
22	H	$(CH_2)_2O(CH_2)_2OH$	H	$(CH_2)_2O(CH_2)_2OH$	H

The following non-limitative examples are to further illustrate the present invention.

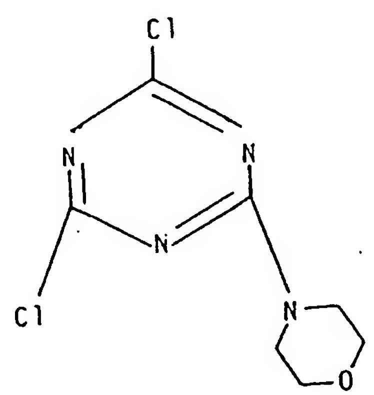
EXAMPLE 1

Into a 2 l-reactor equipped with stirrer, thermometer, dropping funnel, condenser and cooling bath, there are placed 184.5 g of cyanuric acid chloride and 1300 ml of methylene chloride.

While cooling externally, 87.2 g of morpholine and 40 g of sodium hydroxide dissolved in 150 g of water are added simultaneously within 3 hours, keeping the pH between 5 and 7 and the temperature between 0 and 3°C.

The mixture is kept at 0 to 3°C for a further 3 hours and thereafter the aqeuous phase is separated.

Upon distillation of the methylene chloride, 230 g of the intermediate of formula (IX):



(IX)

are obtained in the form of a white crystalline powder having a melting point (m.p.) of 155-157°C and a chlorine content of 30.12% (theor. = 30.21%).

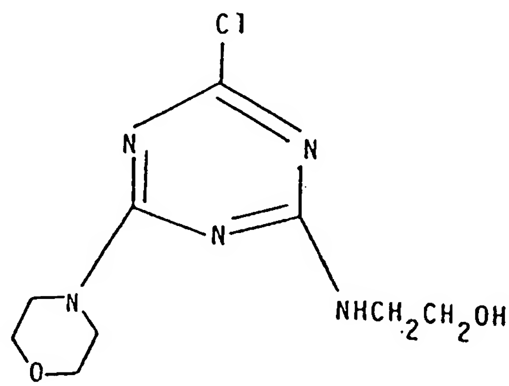
In a 1 l-reactor equipped with stirrer, thermometer, feeding funnel, cooler and heating bath, there are introduced 300 ml of water, 30.5 g of 2-hydroxyethylamine and, under stirring, 117.5 g of the intermediate (IX).

The temperature is gradually raised to 40°C; after 30 minutes the mixture is heated to 45°C and is kept at this temperature for about 3 hours.

Then the temperature is raised to 50°C and within 3 hours a solution of 20 g of sodium hydroxide in 100 ml of water is added.

The resulting mixture is kept at 50°C for a further 2 hours and thereafter is heated to 70°C and is allowed to further react at this temperature for 30 minutes.

After cooling to room temperature, the product formed is filtered off and washed on the filter with water. After drying of the cake in an oven at 100°C, 120.3 g of the intermediate of formula (X):



(X)

are obtained in the form of a white crystalline powder the m.p. of which is 172-173°C and the chlorine content of which is 13.51% (theor.: 13.68%).

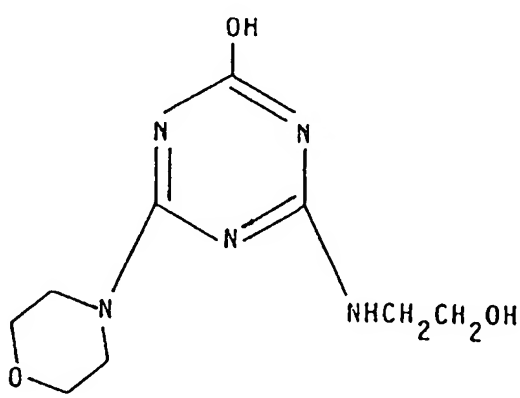
The structure of the intermediates (IX) and (X) was further confirmed by IR spectroscopic analysis.

The same 1 l reactor is charged with 500 ml of water, 103.8 g of the intermediate (X) and 79 g of a 37% by weight solution of hydrochloric acid.

The mass is heated to 90°C and is kept at this temperature for 3 hours. Then the solution is cooled to 50°C and is neutralized by adding 48 g of sodium hydroxide dissolved in 80 ml of water.

The resulting mixture is cooled to 5°C and the product formed is filtered and washed on the filter with cold water.

Upon drying the filter cake in an oven at 100°C, 84.7 g of the product of formula



are obtained in the form of a white crystalline powder; m.p. = 251-253°C.

EXAMPLE 2

Into a 2 l-reactor, equipped as in example 1, 800 ml of methanol, 100 ml of water and 151.2 g of sodium bicarbonate are introduced.

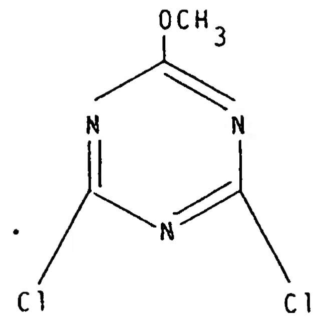
The mixture is cooled to 10°C and then 166 g of cyanuric acid chloride are added.

The temperature is allowed to rise up to 30°C and is kept at this value for about 1 hour, until completion of the carbon dioxide evolution.

The heat of reaction itself is sufficient for maintaining the desired temperature.

Then the reaction mixture is cooled to 5°C and thereafter 800 ml of cold water are added. The product formed is filtered off and washed on the filter with cold water.

Upon drying the cake in an oven under vacum at 60°C, 123.8 g of the intermediate of formula (XI):



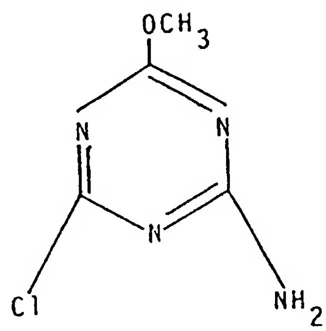
(XI)

are obtained in the form of a white crystalline powder; m.p. = 90-92 °C; chlorine content 39.27% (theor. 39.44%).

In a 1 l-reactor, equipped as in example 1, 400 ml of water and 108 g of the intermediate (XI) are introduced.

While cooling externally to 0-5 °C, 100 g of a 30% by weight aqueous ammonia solution are added within about 1 hour while keeping the temperature at 0 to 5 °C. Then the temperature is allowed to rise spontaneously to room temperature and this value is maintained for 2 hours.

The resulting mixture is cooled to 10 °C and the product formed is filtered off and washed with cold water. By drying the cake in an oven at 100 °C, 82.3 g of the intermediate of formula (XII):



(XII)

are obtained in the form of a white crystalline powder; m.p. >300 °C; chlorine content 21.96% (theor.: 22.12%).

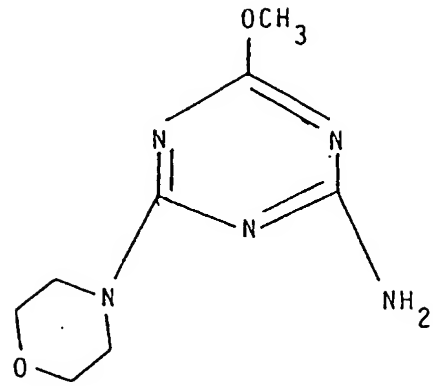
The structure of intermediates (XI) and (XII) was further confirmed by NMR analysis.

In the same 1 l-reactor, but provided with a heating bath, 300 ml of toluene, 80.2 g of the intermediate (XII) and 90 g of morpholine are introduced.

The mixture is heated to 60 to 65 °C and is kept at this temperature for 2 hours; thereafter the mixture is heated to reflux and is kept in this condition for 1 hour.

After having allowed the mixture to cool to room temperature, the product formed is separated by filtration.

The cake is thoroughly washed with water and, after drying, 90.3 g of the product of formula



are obtained in the form of a white crystalline powder; m.p. = 182-184 °C.

EXAMPLE 3

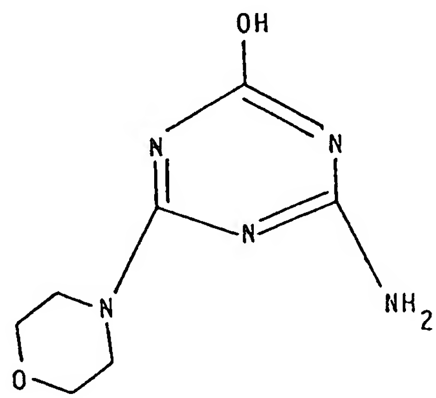
In a 1 l-reactor, equipped as in example 2, 400 ml of water, 63.3 g of the product of example 2 as well as 59.1 g of a 37% by weight hydrochloric acid solution are placed.

The mixture is heated to boiling and kept under reflux for 2 hours.

After cooling to 80 °C, 24 g of sodium hydroxide dissolved in 100 ml of water are added.

The resulting mixture is allowed to cool to room temperature, then the product formed is filtered off and washed on the filter with water.

Upon drying the cake in an oven at 100 °C, 54.7 g of the product of formula:



are obtained in the form of a white crystalline powder; m.p. >300 °C.

EXAMPLE 4

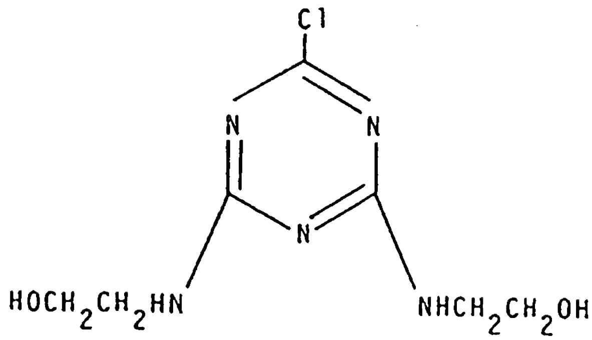
600 ml of water and 184.5 g of cyanuric acid chloride are placed in a 2 l reactor equipped as in example 1.

While externally cooling to 2 °C, 122.5 g of 2-hydroxyethylamine in 100 ml of water are added within 2 hours; during the addition the temperature is allowed to gradually rise up to 5 to 7 °C.

Then the temperature is raised to 20 °C and is kept at this value for 1 hour; thereafter the whole mixture is heated to 35 to 40 °C and 80 g of sodium hydroxide (dissolved in 200 ml of water) are added within about 3 hours.

The reaction mass is heated to 60 °C and is kept at this temperature for 2 hours, whereafter it is cooled to room temperature and the product formed thereby is filtered off and washed on the filter with water.

By drying the cake in an oven at 100 °C, 203.1 g of the intermediate of formula (XIII):



(XIII)

are obtained in the form of a white crystalline powder; m.p. = 188-190 °C; chlorine content 15.33% (theor. 15.20%).

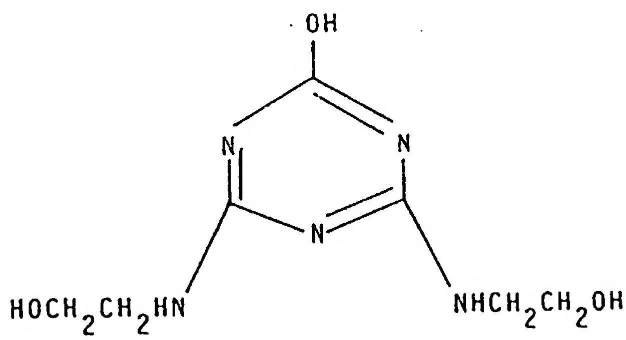
The structure of the intermediate (XIII) was further confirmed by IR spectroscopic analysis.

400 ml of water, 13 g of sodium hydroxide and 70.1 g of the intermediate (XIII) are placed in a 1 l stainless steel reactor equipped as in example 1.

The mixture is then heated to 150 °C and is kept at this temperature for about 10 hours.

Thereafter the reaction mass is cooled to room temperature and the product formed is filtered and washed on the filter with water.

By drying the cake in an oven at 100 °C, 57.7 g of the product of formula

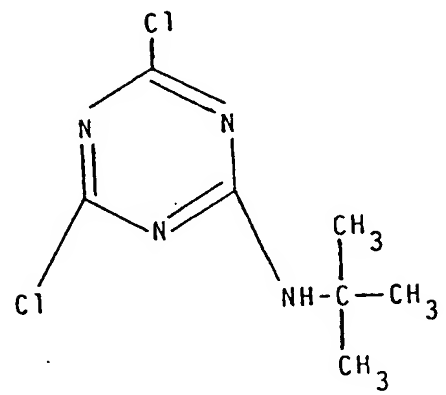


are obtained in the form of a white crystalline powder; m.p. >300 °C.

EXAMPLE 5

1000 ml of methylene chloride, 129.1 g of cyanuric acid chloride, 51.2 g of tert-butylamine and 28 g of sodium hydroxide dissolved in 100 ml of water are placed in the 2 l-reactor described in example 1.

By working as described in example 1, after having distilled off the solvent, 148 g of the intermediate of formula (XIV):



(XIV)

are obtained in the form of a white crystalline powder; m.p. = 129-130 °C; chlorine content 31.87% (theor. 32.13%).

500 ml of chloroform, 110.5 g of the intermediate (XIV) and 30.5 g of 2-hydroxyethylamine, dissolved in 80 ml of water, are placed in a 1 l-reactor equipped as in the preceding examples.

The mixture is heated to boiling and kept under reflux for 3 hours; thereafter, a solution of 20 g of sodium hydroxide in 70 ml of water is added within 2 hours.

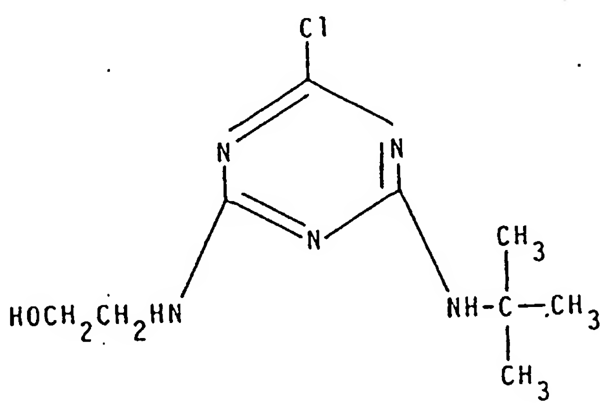
The mixture is kept under reflux for a further hour and then is cooled to room temperature by separating the organic phase.

The solvent is distilled off and the distillation residue is treated with 500 ml of water in the same 1 l-reactor.

The product is heated to 50 to 60 °C until a good dispersion is obtained and thereafter the mixture is cooled to room temperature and the product formed is separated by filtration.

The filter cake is washed with water and dried in an oven at 80 °C.

106.7 g of the intermediate of formula (XV):



(XV)

are obtained in the form of a white crystalline powder; m.p. = 134-135 °C; chlorine content 14.32% (theor. 14.46%).

The structure of intermediates (XIV) and (XV) was further confirmed by NMR analysis.

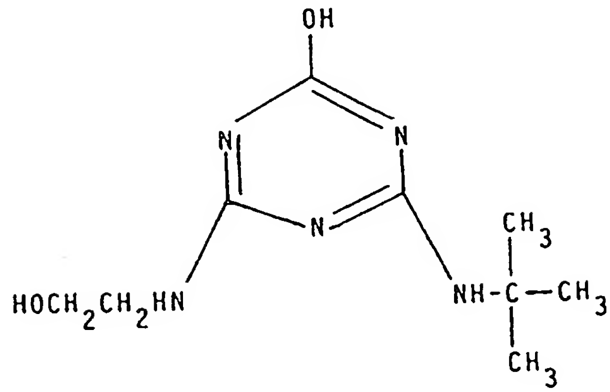
500 ml of water, 98.2 g of the intermediate (XV) and 20.5 g of 96% by weight sulfuric acid are placed in the same 1 l-reactor.

The reaction mass is heated to 85 °C and is kept at this temperature for 2 hours.

Thereafter, 32 g of sodium hydroxide, dissolved in 100 ml of water, are added within 30 minutes.

The resulting mixture is kept at 85 °C for a further 30 minutes and thereafter is cooled to room temperature; the product formed is filtered and washed on the filter.

Upon drying the cake in an oven at 100 °C, 83.2 g of the product of formula



are obtained in the form of a white crystalline powder; m.p. >300 °C.

EXAMPLE 6

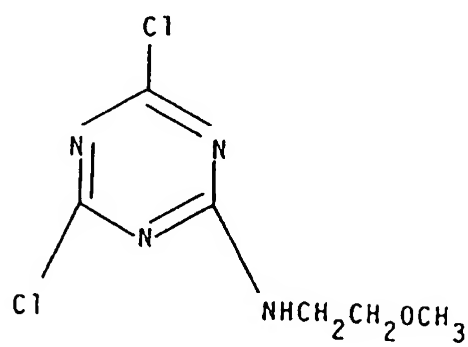
600 ml of water and 184.5 g of cyanuric acid chloride are introduced in a 2 l-reactor equipped as described in example 1.

While cooling from the outside to 0 to 2 °C, 75 g of 2-methoxyethylamine are added within 1.5 hours.

Subsequently, 40 g of sodium hydroxide, dissolved in 250 ml of water, are introduced within 2 hours, always maintaining the temperature at 0-2 °C.

The mass is kept under stirring for a further hour at the same temperature, then the product formed is separated by filtration and washed on the filter with water.

By drying in oven under vacuum at 60 °C, 178.9 g of the intermediate of formula (XVI):



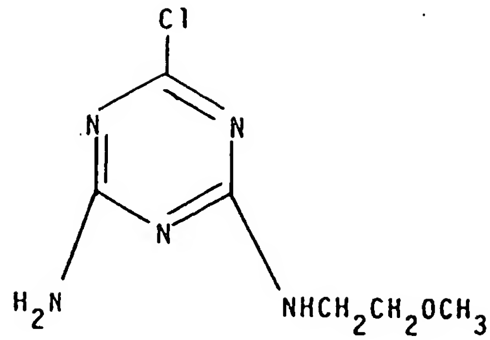
(XVI)

are obtained in the form of a white crystalline powder; m.p. = 73-75 °C; chlorine content 31.68% (theor. 31.84%).

In a 1 l-reactor, equipped as in example 1, there are introduced 85 g of a 30% by weight aqueous ammonia solution, 250 ml of water and 111.5 g of intermediate (XVI).

The mixture is first heated to 40 °C, keeping this temperature for 4 hours, then to 55 °C for 2 hours. Thereafter the reaction mass is cooled to 10 °C and the product formed is filtered and washed on the filter with water.

By drying the cake in oven at 100 °C, 98 g of the intermediate of formula (XVII):



(XVII)

are obtained in the form of a white crystalline powder; m.p. = 195-197 °C; chlorine content 17.21% (theor. 17.44%).

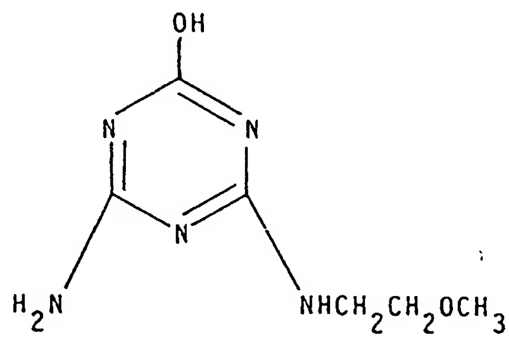
The structure of intermediates (XVI) and (XVII) was further confirmed by IR spectroscopic analysis. 400 ml of water, 81.4 g of the intermediate (XVII) and 42.3 g of a 37% by weight hydrochloric acid are placed in the same 1 l-reactor.

The mixture is heated to 80 °C and is kept at this temperature for 2 hours.

Thereafter, 44.2 g of sodium carbonate, dissolved in 200 ml of water, are added, always at 80 °C.

The reaction mass is then cooled to room temperature and the product formed is filtered and washed on the filter with water.

By drying the cake in an oven at 100 °C, 68.1 g of the product of formula



are obtained in the form of a white crystalline powder having a melting point of >300 °C.

EXAMPLE 7

400 ml of methanol are introduced in a 1 l-reactor equipped as in the preceding examples and, while keeping the temperature at 15 to 20 °C, 17.6 g of sodium hydroxide are added.

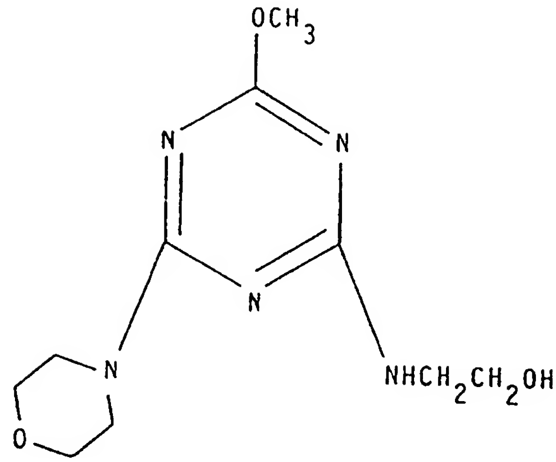
The mixture is kept under stirring until the dissolution of the sodium hydroxide is complete, whereafter 103.8 g of the intermediate (X) are added.

The resulting mixture is heated to boiling and is kept under reflux for 4 hours.

Then most of the solvent (about 350 ml) is distilled off and the residue of the distillation is treated with 200 ml of water.

The product formed is filtered and washed on the filter with water.

By drying the cake in an oven at 100 °C 88.7 g of the product of formula



are obtained in the form of a white crystalline powder; m.p. = 136-138 °C.

EXAMPLES 8 TO 24

By working under conditions analogous to those described in examples 1 to 7, the compounds of general formula (I) listed in Table 2 below are prepared.

TABLE 2

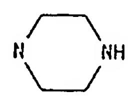
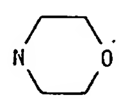
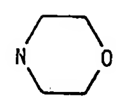
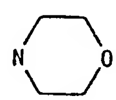
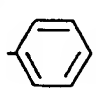
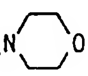
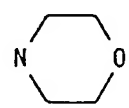
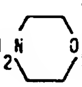
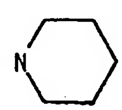
EX. No.	R	R_1—(N—)—R_2		R_3—(N—)—R_4		m.p. (°C)
8	H	$(CH_2)_3OH$	H	$(CH_2)_3OH$	H	>300
9	H	CH_2CH_2OH	H	CH_2-CH=CH_2	H	>300
10	H	$CH_2CH_2OCH_3$	H	$CH_2CH_2OCH_3$	H	>300
11	H			H	H	>300
12	H	CH_2CH_2OH	CH_3	CH_2CH_2OH	CH_3	207-209
13	H	$(CH_2)_5OH$	H	H	H	254-257
14	H					> 300
15	n-C_4H_9			H	H	109-113
16	H	$CH_2CH(OH)CH_3$	H	$CH_2CH(OH)CH_3$	H	>300
17		CH_2CH_2OH	H	CH_2CH_2OH	H	238-242

TABLE 2 (continued)

EX. No.	R	R_1—(—N—)—R_2		R_3—(—N—)—R_4		m.p. (°C)
18	H	CH_2CH_2OH	CH_2CH_2OH	CH_2CH_2OH	CH_2CH_2OH	200-203
19	H	$CH_2CH_2CH_2$	H	H	H	> 300
20	CH_2-CH=CH_2			H	H	149-152
21	H	$CH_2CH_2OCH_3$	H	CH_2CH_2OH	H	250-253
22	CH_2CH_2	CH_2CH_2OH	H	CH_2CH_2OH	H	227-231
23	H	$CH_2CH_2OCH=CH_2$	H	H	H	> 300
24	H	CH_2CH_2OH	H			>300

EXAMPLES 25 TO 73

The tests reported in the following Tables 3 and 4 relate to polymeric compositions containing the products of general formula (I) prepared according to the preceding examples.

Specimen having the shape of small slabs of about 3 mm of thickness were prepared by moulding mixtures of granular polymer and additives on a MOORE platen press, with a moulding cycle of 7 minutes and a moulding pressure of about 40 kg/cm^2.

On the slab specimen thus obtained the level of self-extinguishment was determined by measuring the oxygen index (L.O.I. according to ASTM D-2863-77 in a Stanton Redcroft instrument) and by applying the "Vertical Burning Test" which makes it possible to classify the material according to three rating levels (94 V-O, 94 V-1 and 94 V-2) according to the UL 94 standard (issued by Underwriters Laboratories - U.S.A.).

In Table 3 the values which were obtained by using an isotactic polypropylene in flake form and having

a melt flow index of 12 and an insoluble fraction in boiling n-heptane of 96% by weight are reported.

In Table 4 there are reported the values which were obtained by using low-density polyethylene in granular form with a melt flow index of 7; polystyrene granules containing 5% by weight of butadiene rubber and having a melt flow index of 9; thermoplastic polyurethane granules, of polyester grade (ESTANE 54600 ® by Goodrich) or of polyether grade (ESTANE 58300 ® by Goodrich), having respective specific gravities of 1.19 and 1.10 g/cm^3; an ethylene-propylene elastomeric copolymer containing 45% by weight of propylene; and an acrylonitrile-butadiene-styrene terpolymer having a specific gravity of 1.06 g/cm^3 and a melt flow index of 1.6, and containing about 40% of each acrylonitrile and styrene and 20% butadiene.

TABLE 3

Example No.	Product Example No.	PARTS BY WEIGHT Product	PP (1)	AO (2)	APP (1)	L.O.I. (ASTM D2863)	UL 94 3 mm
25	1	7,4	73	1	18.6	33,8	V0
26	2	8,3	70	1	20.7	34,6	V0
27	3	16,0	75	1	8.0	29.5	V1
28	4	4,2	78	1	16.8	31.8	V0
29	5	6.9	75	1	17.1	31,4	V1
30	6	8.0	75	1	16.0	33,3	V0
31	7	8.3	70	1	20.7	34,7	V0
32	8	7.2	70	1	21.8	37,1	V0
33	9	8.3	70	1	20.7	33.8	V0
34	10	7.2	70	1	21,8	36.5	V0
35	11	6.9	75	1	17,1	30.8	V0
36	12	6.8	75	1	17.2	31,4	V0
37	13	9,7	70	1	19.4	32.1	V1
38	14	6,8	75	1	17.2	34.6	V0
39	15	8.3	70	1	20.7	31.8	V1
40	16	6.0	75	1	18,0	34.7	V0
41	17	8.3	70	1	20.7	32,3	V1
42	18	6.0	75	1	18.0	34.3	V0
43	19	12.0	75	1	12,0	31,3	V0
44	20	8.3	70	1	20.7	32,4	V0
45	21	6.8	75	1	17,2	32.7	V0
46	22	8,3	70	1	20.7	34.7	V0
47	23	8.0	75	1	16.0	33.6	V0
48	24	7,2	70	1	21.8	32,9	V0
49	3	8.0	75	1	16.0*	31,5	V0
50	6	6.0	75	1	18.0*	34,1	V0
51	4	6.0	75	1	18.0(3)	30,5	V0
52	16	6.8	75	1	17.2(4)	31.8	V0

(1) PP = polypropylene
APP = ammonium polyphosphate
Exolit® 422 (Hoechst)

*	APP	=	ammonium polyphosphate microincapsulated with melamine-formaldehyde resin Exolit ® 462 (Hoechst)

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(2)	AO	=	antioxidant Mixture consisting of two parts of dilauryl thiopropionate and 1 part of pentaerythritol tetra[3-(3,5-di-tert-butyl-4-hydroxyphenyl)-propionate]

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(3)	monoammonium salt of 1-aminoethane-1,1-diphosphonic acid

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(4)	monoammonium salt of 1-hydroxyethane-1,1-diphosphonic acid.

TABLE 4

Ex. No.	Supp. Polymeric (1) (1)	Product Example No.	PARTS BY WEIGHT				L.O.I. (ASTM-D2863)	UL94 3 mm
			Polymer	Product	AO (2)	APP (1)		
53	LDPE	1	70	7.2	1	21.8	30.0	V1
54		4	65	8.5	1	25.5	35.7	V0
55		16	60	9.0	1	30.0	40.7	V0
56		18	65	8.5	1	25.5	32.1	V1
57	HIPS	3	65	11.3	1	22.7	29.2	V1
58		4	65	13.6	1	20.4	32.7	V0
59		8	65	11.3	1	22.7	31.5	V1
60		16	65	8.5	1	25.5	30.5	V0
61		18	63	12.0	1	24.0	31.8	V0
62		4	64	8.8	1	26.2	29.8	V0
63	PU (ester)	1	70	7.2	1	21.8	33.8	V0
64		3	70	7.2	1	21.8	35.2	V0
65		4	70	8.3	1	20.7	36.1	V0
66		10	70	7.2	1	21.8	34.4	V0
67		16	70	8.3	1	20.7	34.5	V0
68	PU (ether)	3	65	8.5	1	25.5	28.9	V0
69	PP/PE	1	65	8.5	1	25.5	30.4	V1
70		4	65	9.7	1	24.3	32.5	V0
71		16	65	8.5	1	25.5	30.8	V0
72	ABS	4	65	13.6	1	20.4	29.7	V0
73		16	65	13.6	1	20.4	28.5	V0

(1) APP = ammonium polyphosphate- Exolit® 422 (Hoechst)

LDPE = low density polyethylene

HIPS = polystyrene containing 5% of butadiene rubber

PU (ester)	=	polyurethane polyester
PU (ether)	=	polyurethane polyether
PP/PE	=	propylene-ethylene copolymer
ABS	=	acrylonitrile-butadiene-styrene terpolymer
(2) AO	=	antioxidant Mixture consisting of 2 parts of dilauryl thiopropionate and 1 part of pentaerythritol tetra[3-(3,5-di-tert-butyl-4-hydroxyphenyl)-propionate]
(3)	=	monoammonium salt of phenylphosphonic acid.

EXAMPLE 74 (comparison example)

By working according to the procedure used in examples 25 to 52, but using as nitrogen containing compound 2,4-diamino-6-hydroxy-1,3,5-triazine, the composition specified hereinafter is prepared:

Polypropylene	72 parts by weight
Antioxidant	1 part by weight
Ammonium polyphosphate	19.3 parts by weight
2,4-diamino-6-hydroxy-1,3,5-triazine	7.7 parts by weight

Using the above mentioned composition specimens are prepared which are subjected to the self-extinguishing tests described above.

The following results are obtained:

L.O.I. = 23.8

UL 94 (3 mm): class B (the specimen burns).

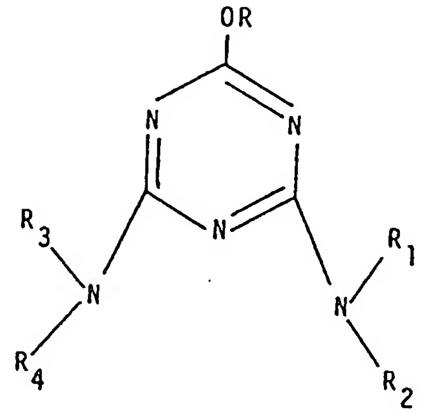
Claims

1. Self-extinguishing polymeric compositions comprising:

    a) from 91 to 40 parts by weight of thermoplastic polymer and/or of polymer showing elastomeric properties;

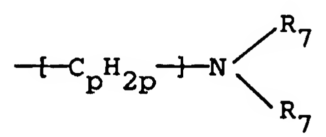
    b) from 6 to 33 parts by weight of one or more compounds selected from ammonium and amine phosphates and ammonium and amine phosphonates;

    c) from 3 to 27 parts by weight of one or more 2,4-diamino-6-hydroxy-1,3,5-triazine derivatives of general formula (I):



(I)

wherein:
R = H; $(C_2\text{-}C_6)$-alkenyl; $(C_6\text{-}C_{12})$-cycloalkyl; $(C_6\text{-}C_{12})$-alkylcycloalkyl; $(C_6\text{-}C_{12})$-aryl; $(C_7\text{-}C_{12}$-aralkyl; and $\{C_nH_{2n}\}X$,
n being an integer of from 1 to 8 and X being selected from H; CN; -O-$(C_1\text{-}C_4)$-alkyl;-O-$(C_2\text{-}C_4)$-alkenyl; $(C_6\text{-}C_{12})$-cycloalkyl; $(C_6\text{-}C_{12})$-alkylcycloalkyl; -O-$(C_6\text{-}C_{12})$-aryl; moieties -$N(R_5)_2$ wherein the groups R_5, the same or different from each other, represent $(C_1\text{-}C_4)$-alkyl or $(C_3\text{-}C_4)$-alkenyl; or the moiety -$N(R_5)_2$ is replaced by an N-heterocyclic radical which optionally contains another heteroatom and is linked to the alkyl chain through the nitrogen atom;
at least one of the groups R_1, R_2, R_3 and R_4 is selected from:
$\{C_mH_{2m}\}O\text{-}R_6$; and



wherein
m = an integer of from 2 to 8;
p = an integer of from 2 to 6;
R_6 = H; $(C_1\text{-}C_8)$-alkyl; $(C_2\text{-}C_6)$-alkenyl; $\{C_qH_{2q}\}OR_8$, wherein q is an integer of from 1 to 4 and R_8 is hydrogen or $(C_1\text{-}C_4)$-alkyl; $(C_6\text{-}C_{12})$-cycloalkyl or $(C_6\text{-}C_{12})$ -alkylcycloalkyl;
the groups R_7, the same or different from each other, represent H; $(C_1\text{-}C_8)$-alkyl; $(C_2\text{-}C_6)$-alkenyl; $(C_1\text{-}C_4)$-hydroxyalkyl; $(C_6\text{-}C_{12})$-cycloalkyl and $(C_6\text{-}C_{12})$-alkylcycloalkyl;
or the moiety -$N(R_7)_2$ is replaced by an N-heterocyclic radical which optionally contains another heteroatom and is linked to the alkyl chain through the nitrogen atom; or in general formula (I) at least one of the moieties -NR_1R_2 and -NR_3R_4 is replaced by an N-heterocyclic radical which optionally contains another heteroatom and is linked to the triazine ring through the nitrogen atom; the remaining groups R_1 to R_4, the same or different from each other, having the meanings mentioned above or being selected from H; $(C_1\text{-}C_{18})$-alkyl; $(C_2\text{-}C_8)$-alkenyl; $(C_6\text{-}C_{16})$-cycloalkyl and $(C_6\text{-}C_{16})$-alkylcycloalkyl, optionally substituted by a hydroxy or $(C_1\text{-}C_4)$-hydroxyalkyl group.

2. Compositions according to claim 1, wherein at least one of the moieties -NR_1R_2 and -NR_3R_4 in general formula (I) is replaced by a heterocyclic radical selected from aziridyl, pyrrolidyl, piperidyl; morpholyl; thiomorpholyl, piperazyl; 4-methylpiperazyl; 4-ethylpiperazyl; 2-methylpiperazyl; 2,5-dimethylpiperazyl; 2,3,5,6-tetramethylpiperazyl; 2,2,5,5-tetramethylpiperazyl; 2-ethylpiperazyl and 2,5-diethylpiperazyl.

3. Compositions according to any one of claims 1 and 2, wherein at least one of the groups R_1 to R_4 in general formula (I) is a group of formula

$\{C_mH_{2m}\}O\text{-}R_6$

wherein m = an integer of from 2 to 4 and
R_6 is hydrogen or $(C_1\text{-}C_4)$-alkyl.

4. Compositions according to any one of claims 1 to 3, wherein the moiety $-N(R_5)_2$ is replaced by a heterocyclic radical selected from pyrrolidyl; piperidyl; morpholyl; thiomorpholyl; piperazyl; 4-methylpiperazyl and 4-ethylpiperazyl.

5. Compositions according to any one of claims 1 to 4, wherein the moiety $-N(R_7)_2$ is replaced by a heterocyclic radical selected from aziridyl; pyrrolidyl; piperidyl; morpholyl; thiomorpholyl; piperazyl; 4-methylpiperazyl and 4-ethylpiperazyl.

6. Compositions according to any one of claims 1 to 3 and 5 wherein R in general formula (I) represents hydrogen or $\{C_nH_{2n}\}X$ wherein
   n is an integer of from 1 to 4 and
   X is hydrogen.

7. Compositions according to any one of claims 1 to 6, wherein the ammonium phosphates (b) have the general formula $(NH_4)_{n+2}P_nO_{3n+1}$ wherein n is an integer of at least 2; or have the general formula $(NH_4PO_3)_n$ wherein n ranges from 50 to 500.

8. Compositions according to any one of claims 1 to 7, wherein the amine phosphates (b) are selected from dimethylammonium phosphate; diethylammonium phosphate; ethylenediamine phosphate; melamine ortho-phosphate; melamine pyrophosphate; and mixtures thereof.

9. Compositions according to any one of claims 1 to 8, wherein the ammonium phosphonates (b) are the mono- and polysubstituted ones and are selected from salts derived from mono- and polyphosphonic acids.

10. Compositions according to any one of claims 1 to 9, wherein the polymer (a) is selected from acrylonitrile/butadiene/styrene (ABS) terpolymers; styrene/acrylonitrile (SAN) copolymers; polyurethanes; poly(ethyleneterephthalate); poly(butylene terephthalate); polyamides; and polymers and copolymers of olefins of general formula $R'\text{-}CH=CH_2$ wherein R' is hydrogen, a $(C_1\text{-}C_8)$-alkyl or $(C_6\text{-}C_{10})$-aryl group, particularly:
    (1) isotactic or prevailingly isotactic polypropylene;
    (2) HDPE, LLDPE and LDPE polyethylene;
    (3) crystalline copolymers of propylene and minor amounts of ethylene and/or other alpha-olefins, such as butene-1, hexene-1, octene-1 and 4-methylpentene-1;
    (4) heterophasic compositions comprising (A) a fraction of propylene homopolymer or of propylene copolymers as specified under (3) and (B) a copolymeric fraction composed of elastomeric copolymers of ethylene and an alpha-olefin, optionally containing minor amounts of a diene, wherein the alpha-olefin is preferably selected from propylene and butene-1;
    (5) elastomeric copolymers of ethylene and alpha-olefin(s) optionally containing minor amounts of diene.

11. Molded articles, obtained from compositions according to any one of claims 1 to 10.